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DIFFERENCE FREQUENCY GENERATION BY NONLINEAR OPTICS METHODS

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ABSTRACT. Theoretical investigation of the conversion of optical emission from powerful solid-state lasers into the far infrared region by means of nonlinear polarization with the generation of a small difference frequency. It is shown that the spatial divergence of the input radiation is very significant, and the surface brightness of the output face of the crystal is determined at the difference frequency. Attention is given to cases of emission under conditions of phase synchronization and without synchronization. In the first case, the emitted radiation is concentrated within an interval of angles up to several degrees, while in the second case it is distributed in practically all directions with the crystal's edge surface brightness weakly dependent on the output angle. It is shown that the use of giant-pulse lasers can result in emission in the far infrared at power levels of 10 W or more. A70-38741

One of the important applications of nonlinear optics is the transformation of energy from laser emission to the frequency scale. This transformation has been successfully carried out both with increased and decreased frequencies [1]. There is particular interest in the generation of a difference frequency (DF) in the far infrared region during excitation by emission of powerful solid-state lasers of the optical range, particularly in connection with the problem of producing sources of coherent emission in this region. This generation was done experimentally by the authors of [2 - 4]. /861*

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*Numbers in the margin indicate the pagination in the original foreign text.

The purpose of this report is to present the theory regarding the generation of small DF, which has several characteristics as compared to the well-studied case when the input and transformed frequencies are of the same order of magnitude.

Method of examination. It is assumed that exciting frequencies $\omega_{1,2}$ are so close that the transformed frequency $\omega = \omega_1 - \omega_2 \ll \omega_{1,2}$. The main characteristic of this generation is the anomalously large influence of the spatial dispersion [5]. The finite nature of the frequency spectrum under real conditions is not as important, and we shall not take it into account.

We shall discuss the following characteristics of emission of solid-state lasers. It usually contains a large number of slightly synchronized modes [6], and, therefore, statistically independent transverse and longitudinal modes. The spatial dispersion of real laser beams is not large, as a rule. This provides a basis for selecting a limited observation region (in terms of length — on the order of the crystal thickness l), whose dimensions are small as compared with the distance to the equivalent phase center. We can disregard the spatial nonuniformity of the emission field, assuming that it is approximately uniform in the sense that the correlation functions of the field components at different points depend only on the distance between these points. Similarly to the steady state condition [7], to do this it is necessary that the correlation moment of different Fourier components of the field $E_0(\vec{x})$, $E_0(\vec{x}')$ be proportional to $\delta(\vec{x}+\vec{x}')$, i.e., $\frac{c}{4\pi} n_0 \langle E_0(\vec{x}) E_0(\vec{x}') \rangle = I_0(\vec{x}) \delta(\vec{x}+\vec{x}')$ (1).

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(1) The field is assumed to be approximately linearly polarized, or consisting of several beams of different polarization, so that this relationship is valid for each beam individually, and $E_0(\vec{x}) = eE(\vec{x})$; e — polarization unit vector. In view of the slight dispersion of the exciting emission, the difference in the polarization of individual components and values of the refractive index n_0 is very small within the limits of each beam. In addition it is assumed that the medium is slightly anisotropic, so that the deviation of the emission field from a transverse field can be ignored.

Thus, the Poynting-Umov vector $S_0 = \int s_0 I_0(\vec{\varkappa})\, d^3\varkappa$ (s_0 — the unit vector along $\vec{\kappa}$) has the same form as for a family of continuously distributed, noncoherent plane waves. Therefore, this approximation may be called the approximation of noncoherent plane waves.

In the case of nonlinear optics effects of the second order of magnitude, the intensity of the transformed emission is determined by the correlation moments of the fourth order of the Fourier components of the exciting field, and by the functions which describe the generation of polarization by individual induced plane waves. The field E produced at the ouput of the medium (in a vacuum) by the nonlinear portion of its polarization represents a linear function of the latter, so that each induced plane wave of polarization $P(q)$ produces a definite plane wave $E(k)$, and $E(k_x, k_y, k_z) = R(q_x, q_y, q_z) P(q_x, q_y, q_z)$, and $q_z \geqslant 0$ $k_z = \sqrt{(\omega/c)^2 - k_\tau^2}$, $k_{x,y} = q_{x,y}$, $k_\tau^2 = k_x^2 + k_y^2$. The explicit form of the coefficients $R(q)$ may be readily found by using the well-known results of calculations of generation by plane waves [8]. The transformed emissions may also be represented by the superposition of noncoherent plane waves, and the Poynting-Umov vector S in a vacuum has the following form for it:

$$S = \int s I(k)\, d^3k, \quad I(k) = \frac{4\pi}{c} |dR(q)|^2 A(q), \tag{1}$$

$$A(q) = 2 \int I_0(q - \vec{\varkappa})\, I_0(\vec{\varkappa})\, d^3\varkappa + I_0^2 \delta(q), \quad I_0 = \int I_0(\vec{\varkappa})\, d^3\varkappa. \tag{2}$$

Here s is the unit vector along the wave vector of the corresponding plane wave in a vacuum k; d — length of the vector with the components $d_l = \chi_{ijk} \times$ $(\varkappa_{01}, \omega_1; \varkappa_{02}, \omega_2) e_j(\varkappa_{01}, \omega_1) e_k(\varkappa_{02}, \omega_2)$; χ_{ijk} — tensor components of nonlinear polarizability, whose spatial dispersion throughout the beam can be disregarded. The second term in the expression for $A(q)$ is important only in the case of optical detection [9], which we shall not examine.

There is great interest in a luminous flux B of radiation of frequency ω emitted by a unit surface of crystal in all directions (i.e., surface

luminosity), equal to B = (S, N) = $\int (s, N) I(k) d^3k$ (N — normal to the back face, going from the crystal). The surface brightness B_Ω connected with B by the relationship $B = \int B_\Omega d\Omega$ ($d\Omega$ — element of a solid angle in the space of the vectors k) is also of interest. The quantity B_Ω may be found as

$$B_\Omega = k^2 \cos^2\theta \int I(k_x, k_y, q_z)\, dq_z, \quad k_x = k \sin\theta \cos\varphi,$$
$$k_y = k \sin\theta \sin\varphi. \tag{3}$$

To perform specific calculations, it is necessary to determine the function $I_0(\vec{\varkappa})$, which is determined by the Fourier transformation of the correlation function of the exciting emission $\Gamma(r) = \langle E_0(r_1) E_0(r_2) \rangle$, $r = r_1 - r_2$. Unfortunately, the explicit form of correlation functions in the case in which we are interested is unknown. The tentative form of the function $\Gamma(r)$ can be approximated as follows. We shall assume that the length of the crystal l is less than the length of the longitudinal (axis of the beam parallel to the axis z) coherence $l_\parallel \sim c(\Delta\omega)^{-1}$ ($\Delta\omega$ — half width of the spectrum of exciting emission). We may then set $\Gamma(r) = \Gamma_\perp(r_\perp) \cos \varkappa_z z$, $r_\perp = (x^2 + y^2)^{1/2}$. With respect to the function $\Gamma_\perp(r_\perp)$ which describes the correlation in the plane of the transverse cross section, we may obtain specific information about it from the experimental data in [10]. Within the limits of experimental accuracy /863 ($\sim$ 20%), the values of the correlation functions found from experiments may be approximated by the expression $\Gamma_\perp(r_\perp) = C_1 \exp(-ar_\perp^2)$ for a = $5.4 \cdot 10^4$ cm^{-2} and C_1 = const. Other approximation variations are possible. However, final results for the generation of DF are not very sensitive to its explicit selection.

Performing the Fourier transformation of the function $\Gamma(r)$, we obtain the following result

$$I_0(\vec{\varkappa}) = I_0^{(+)}(\vec{\varkappa}) + I_0^{(-)}(\vec{\varkappa}), \quad I_0^{(+)}(\vec{\varkappa}) = C\delta(\varkappa - \varkappa_0) \exp(-\beta 0_\varkappa^2),$$
$$I_0^{(-)}(\vec{\varkappa}) = I_0^{(+)}(-\vec{\varkappa}),$$
$$\beta = (\Delta\theta_\varkappa)^{-2}, \quad C = \frac{\beta}{2\pi\varkappa_0^2} I_0, \quad \varkappa_{01,2} = \frac{\omega_{1,2}}{c} n_{01,2}, \quad I_0(-\vec{\varkappa}) = I_0(\vec{\varkappa}). \tag{4}$$

These formulas completely determine the intensity of generation and its spatial distribution. We shall use them later to examine the generation of small DF, studying individual cases of the presence and absence of phase synchronism. It is assumed that the axes of the beams are perpendicular to the crystal faces.

Case of synchronism. Let us first discuss the question of the dispersion of exciting emission. As already has been pointed out in [5], the bundle of vectors q for waves of the nonlinear portion of polarization by DF is very wide (on the order of several tens of degrees). However, only its small part corresponds to conditions of synchronism. If we take the specific components $\vec{\kappa}_1$ and $\vec{\kappa}_2$ of both exciting beams and designate the angle between them by α (see the figure), then — as may be readily shown — the synchronism region is covered by the opening $\alpha \sim (2\omega\mu c/l)^{1/2}/n_{01}\omega_1$ (μ — refractive index at the frequency ω). Let us make a numerical estimate, assuming that $\omega_{1,2}$ are the frequencies of emission of the lines R_1 and R_2 of a ruby laser ($\lambda_{1,2} = 692{,}9$ and 694.3 Hm [11, 12], λ = 0.34 mm), and the generation occurs in a quartz plate ($n_{01,2} \simeq 1{,}54$, $\mu = 2{,}1$) c $l = 1$ cm. We have taken $\alpha \sim 10^{-4}$, i.e., at least an order of magnitude smaller than the angular dispersion ($\beta^{-1/2} \sim 10^{-3}$—$10^{-2}$) of customary solid state lasers [12]. Since the wings caused by the synchronism conditions can be disregarded, we can conclude that the dispersion greatly (by several orders of magnitude) lowers the generation output.

We find $\theta' \sim 10^{-1}$ from the condition $\sin\theta' = \sin\alpha \dfrac{\varkappa_2}{q}$ (see the figure), so that the dispersion of the generated emission is significantly greater than the input emission. In addition, the angles characterizing the deviation from the general axis of spatially shifted exciting beams of all three vectors $\vec{\varkappa}_1$, $\vec{\varkappa}_2$ and q in a crystal are small. With allowance for this, performing calculations $A(q)$ (2) $c I_0(\vec{\varkappa}) = I_0^{(1)}(\vec{\varkappa}) + I_0^{(2)}(\vec{\varkappa})$, we find

$$A(q) = A^{(+)}(q) + A^{(-)}(q), \quad A^{(+)}(q) = L\delta[q - \varphi(\theta_q)]\exp(-\beta'\theta_q^2),$$

$$\varphi(\theta_q) = q_0\left(1 + \frac{\varkappa_{02}}{\varkappa_{01}}\frac{\theta_q^2}{2}\right),$$

(equations continued)

$$A^{(-)}(q) = A^{(+)}(-q),\ \beta' = \beta\left(\frac{q_0}{\varkappa_{01}}\right)^2,$$

$$q_0 = \varkappa_{01} - \varkappa_{02},\quad L = \frac{\beta}{2\pi\varkappa_{01}^2} I_{01} I_{02}, \tag{5}$$

$$I_{01,2} = \int I_0^{(1,2)}(\vec{\varkappa})\, d^3\varkappa;$$

where θ_q is the angle between q and the axis z. Since $q_0 \ll \varkappa_{01}$, then $\beta' \ll \beta$. Under real conditions $\beta' \sim 1 - 10^{-2}$. Therefore, the factor $\exp(-\beta'\theta_q^2)$ in the synchronism region cannot change, and the agreement between $I(q)$ and θ_q is determined by $g(\tau l) = \left(\frac{\sin \tau l/2}{\tau l/2}\right)^2$, which determines the generation by plane waves and is contained in $|dR(q)|^2 = (\omega l/c)^2 F \cdot g(\tau l)$. Here F is a known constant, whose explicit form depends on the specific geometry of the experiment. In addition, $\tau = (q - q_c, N)$; $q_c = \frac{\omega}{c}\vec{\mu}$ is the wave vector of a natural wave at the frequency ω ($q_{cx} = q_x$, $q_{cy} = q_y$), whose change can be disregarded within the limits of the synchronism region due to anisotropy (if we are speaking about an extraordinary wave of DF). Taking the fact into account that, according to (5), $q = \varphi(\theta_q)$ for $q_z \geqslant 0$ or $\varphi(\pi - \theta_q)$ for $q_z < 0$, we find, respectively[2]

$$\tau_+ = q\cos\theta_q - \sqrt{q_0^2 - q^2\sin^2\theta_q} \simeq q_0 \cdot \frac{\theta_q^2}{2},$$

$$\tau_- = q\cos\theta_q + i\sqrt{q_0^2 - q^2\sin^2\theta_q} \simeq -q_0\,\frac{(\pi - \theta_q)^2}{2}.$$

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(2) Taking into account anisotropy in the case of an extraordinary wave of DF (for exciting waves, in view of the small interval of angles corresponding to synchronism, it is unimportant) would lead to $\tau = q_0(\theta_q^2/2 + \nu\theta_q)$, where in the case of real anisotropy [13] $\nu \sim 10^{-2}$. Therefore, $\nu\theta_q \ll \theta_q^2$ in a large section of the interval of angles θ_q in the synchronism region. Thus, in the case of small DF — in contrast, for example, to the case of the second optical harmonics [14] — the decrease in τ with θ_q is determined primarily not by anistropy, but a change in the length of the vector $q = \vec{\varkappa}_1 - \vec{\varkappa}_2$.

Now we may calculate B_Ω. We obtain $B_\Omega = \frac{2\pi}{c}(2\omega kl/c)^2 FLg(\tau_0 l)$, $\tau_0 = q_0 0^2/2\mu^2$; θ — angle formed by the vector k to the axis z in a vacuum. The total luminous flux $B = B_0 f$; $f = \pi\beta'/q_0 l$; $B_0 = \frac{8\pi}{c}\left(\frac{\omega l}{c}\right)^2 FI_{01}I_{02}$. Here B_0 is the luminous flux generated by parallel plane waves under conditions of precise synchronism. Thus, allowance for the dispersion leads to a decrease in the generation with respect to the factor f, which as a rule is $\sim 10^{-3} - 10^{-1}$.

The DF generation was performed in [2] in quartz under synchronism conditions for lines with λ_1 = 1060.0 NM and λ_2 = 1071.5 NM, which are present in the emission of a neodymium laser. At frequencies of ω_1 and ω, extraordinary waves are excited and at a frequency of ω_2 — ordinary waves. For the case of the experimental geometry in [2], $F = \left\{[4\pi/n_1(1+\mu)]\cos\gamma\left(\chi_{11}\cos\gamma + \frac{\chi_{14}}{2}\sin\gamma\right)\right\}^2$, where $\gamma = 56°$ is the angle between the incident beam and the optical axis of the crystal. A comparison of theory with experiment is made more difficult by the fact that in [2] the values of the necessary parameters are not given. Therefore, only a tentative calculation is possible. The value of χ is /865
identical with the value of the nonlinear susceptibility for optical detection [9, 15], which can be estimated from electro-optical coefficients [16]. We obtain $\chi \simeq 0{,}7\cdot 10^{-8}$ cgs (electrostatic system units). Setting λ = 0.01 cm, $I_{01.2}$ = 50 kW/cm^2, l = 1 cm, the cross section of a laser beam S = $2\cdot 10^{-2}$ sm^2; $\Delta\theta_q = \beta^{-1/2} = 3\cdot 10^{-3}$, we find P_g = BS $\sim 10^{-7}$ W, which coincides with the value of $P_g \sim 5\cdot 10^{-7}$, which is given in [2].

Generation outside of the synchronization conditions. Keeping in mind the comparison with experiments [4], we shall perform the investigation with respect to the specific case of a cubic crystal (without an inversion center), cut so that one of the axes C_2 lies in the plane of the face, and the two other axes form the angles ψ and $\frac{\pi}{2} - \psi$ with the normal to the faces.

The absence of synchronism leads to the absence of selected directions of generation. Since waves of the nonlinear portion of polarization are practically propagated in all directions with $0 \leqslant \theta_q \leqslant \frac{\pi}{2}$ in the case of real values of the dispersion of exciting emission, the same thing is valid in a

given case with respect to DF emission. Therefore, in determining the contraction $A(q)$ (2), it must be assumed that the angle θ_q is small, in spite of the small angles of the dispersion of the exciting emission. Finding $A(q)$ with allowance for this fact and using the Expression (3) for $I_{01,2}(\vec{\varkappa})$, just as previously, and calculating $dR(q)|^2$ according to the well-known method [8], from (3) we find

$$B_\Omega = \frac{128\pi^2\chi^2\xi^2\cos^2\theta\beta_1\beta_2 I_{01}I_{02}}{cn_0^2(\beta_1+\beta_2)} G(\theta)\exp\left(-\beta_1\frac{\xi^2\sin^2\theta}{n_0^2}\right),$$

$$G(\theta) = |dR(q)|^2_{q_z=q_0} = \frac{\varepsilon A_1\sin^2 2\psi + A_2\cos^2 2\psi\sin^2\theta + 1\sqrt{\varepsilon}A_3\sin 4\psi\sin\theta}{\varepsilon(1+1\sqrt{\varepsilon}\cos\theta)^2(b+\cos^2\theta)^2},$$

$$\xi = \frac{\omega_1}{\omega},\ n_0 = n_{01}\simeq n_{02},\ \varepsilon = \varepsilon(\omega),$$

$$A_1 = \alpha^2 + 4\varepsilon - 4\sqrt{\varepsilon}\alpha\rho,\ A_2 = \alpha^2 + 4n_0^2 - 4n_0\alpha\rho,$$

$$A_3 = \alpha^2(1-2\rho) + 4n_0\sqrt{\varepsilon},$$

$$\alpha = n_0 + \sqrt{\varepsilon},\quad \rho = \cos\frac{\omega}{c}(\sqrt{\varepsilon}-n_0)l,\quad b = \varepsilon - n_0^2 - 1, \tag{6}$$

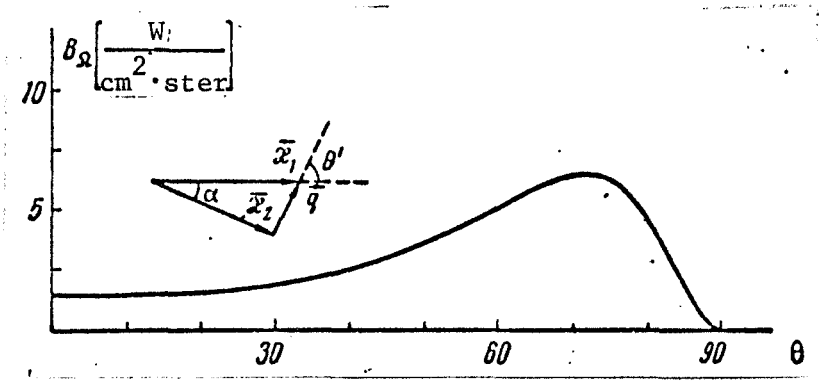
where χ is the value of the tensor component χ_{ijk} which is different from zero. Finally, we have

$$B = \frac{256\pi^3\chi^2\beta_1\beta_2\xi^2 I_{01}I_{02}}{cn_0^2\varepsilon(\beta_1+\beta_2)}\left[(\varepsilon A_1\sin^2 2\psi + A_2\cos^2 2\psi)\int_0^1 u(x)x^2dx - \right.$$

$$\left. - A_2\cos^2 2\psi\int_0^1 u(x)x^4dx + 1\sqrt{\varepsilon}A_3\sin 4\psi\int_0^1 u(x)x^2\sqrt{1-x^2}dx\right],$$

$$u(x) = [(1+\sqrt{\varepsilon}x)(b+x^2)]^{-2}. \tag{7}$$

These formulas are the desired formulas. The figure shows the dependence of $B_\Omega(\theta)$ as applied to the DF generation as observed in [4] on the lines /866 R_1 and R_2 of a ruby laser in the crystal ZnTe (T_d symmetry). The values of the parameters are



Surface brightness of output face ZnTe at frequency ω. Above is the vector diagram for specific components $\vec{\varkappa}_{1,2}$ of exciting beams and the vector $q = \vec{\varkappa}_1 - \vec{\varkappa}_2$ of the polarization wave corresponding to them.

$$\chi = 7{,}3 \cdot 10^{-6} \text{ cgs units}, \; n_0 = 2{,}97, \; \mu = 3{,}18,$$

$$\lambda = 0{,}34 \text{ мм}, \; \xi = 2 \cdot 10^{-3},$$

$$l = 1{,}7 \text{ мм}, \; P_{01,2} = 1 \text{ MW}, \; \Delta\theta_\varkappa = \beta_{1,2}^{-1/2} = 3{,}5 \cdot 10^{-3}, \; \psi = \pi/4.$$

As can be seen, the DF emission is distributed in space more or less isotropically (i.e., the output face of the crystal emits at DF like a Lambert source). The slight rise in the region of large angles is related to the approximation to non-colinear synchronism, which occurred at the angle $\theta_q \approx 22°$ in a crystal. However, complete internal reflection occurs even earlier.

Assuming S = 0.2 mm^2 [4], we find the total intensity of the generated emission P_g = BS = 6 W. The measurements in [4] were made also for a somewhat different value of the dispersion $\Delta\theta_\varkappa = 10^{-2}$. For this case P'_g = 0.8 W. In making a comparison with experiments, it must be kept in mind that under experimental conditions only a tenth of the total intensity of output emission in a

cone with the aperture angle $2\theta_{max} = 40°$ is detected. Performing integration of B_Ω with respect to θ from 0 to 20°, we obtain $P_g = 0.7$ W and $P'_g = 8 \cdot 10^{-2}$ W. These values are in reasonable agreement with the experimental value $\sim 10^{-2} - 10^{-3}$.

The values of P_g obtained in [4] are rather high. However, they do not entirely exhaust all the possibilities of the method. The DF produced in a crystal has been reduced twice by approximately one order of magnitude. This was first due to the total inner reflection: only part of the emission, which is contained in a cone with the aperture angle $2\theta_q = 36°$ within the crystal, passes outside the crystal, and the synchronism direction is not included in it. Therefore, it would be more advantageous to prepare a nonlinear element in the form af a hemisphere, and not a plane-parallel plate, since this would improve the output conditions at the back face. At the output of the crystal, there is a repeated limitation on the angle at which the emission leaving the surface is recorded. There are probably anisotropic semiconductors and dielectrics with large χ, in which collinear synchronization is possible. When the suitable crystals are used, a certain increase in $P_{01.2}$ is permissible. Apparently, generation intensities of $\sim$ 10 W and greater are possible. Further /867 experimental studies in this direction are very desirable.

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